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EVAPORATION AND DISPERSION OF HAZARDOUS MATERIALS

Final Report

Grant No. AFOSR 78-3559

by

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EVAPORATION AND DISPERSION OF HAZARDOUS MATERIALS

Introduction:

This work was undertaken in order to develop a state-of-the-art predictive model for the evaporation and dispersion of hazardous liquids, especially hydro-zine family fuels and propellants, which might be accidentally spilled.

The importance of the problem has been adequately shown previously (1, 2).

Some earlier work by Ille and Springer (3) approached the problem from the standpoint of pure materials. However, the materials of particular interest here are
not always transported or used as pure materials, hence in this work it has been
necessary to incorporate modifications of the earlier work (3) to include mixture
behavior.

An additional consideration has been the incorporation of model elements relating to the reactions of the hazardous materials with the environment, especially water absorption from the air, carbon dioxide absorption from the air and decay of the evaporated material during its transport down wind.

Since the users of this predictive model will ordinarily wish to define the down-wind area wherein a hazard would exist in the case of a spill, the predictive scheme includes down wind dispersion and hazard corridor definition.

All of the several model elements have been assembled and combined in a FORTRAN computer program which will compute evaporation rates and limits of the hazardous region, that is, the maximum distance away from the centerline where a hazard would exist as well as the maximum distance down wind that a hazard would exist. Depending upon the input data supplied, the program will also predict the amount of water that would be required for sufficient dilution to eliminate the hazard, the effects of changing variables (as for example, the effects of changes in wind speed, or in air temperature or in atmospheric stability), and the effect of the passage of time

(up to twenty-four hours) if weather conditions are updated hourly. In the latter case, allowances are made for composition changes in the remaining pool because of preferential evaporation of components, possible absorption of water from the atmosphere, and absorption of (and reaction with) carbon dioxide from the atmosphere.

A number of computations were made in order to determine the effects of changes in the several input parameters. The results of these are presented in the form of correction factors which can be used to correct from certain "base cases", and thus provide for rapid field estimation when needed.

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Nature of the Hazardous Materials Considered:

There are three hazardous (toxic) materials of interest in this work; they being: hydrazine, H2NNH2; monomethyl hydrazine (MMH), H2NNHCH3; and unsymetrical dimethyl hydrazine (UDMH), H2NN(CH3)2. Of these materials, as used by or transported by or for the U.S. Air Force only the MMH is in pure form. The UDMH is used as an equal parts (by mass) mixture with hydrazine. This mixture is sometimes referred to as "Aerozine 50". Hydrazine is also used in a water solution of 70 percent (by mass) hydrazine.

The vapor phase toxicity of these materials has been defined (4), and the Short Term Public Exposure limits (STPL) are given as 7.0 mg/m³, 2.8 mg/m³ and 38.2 mg/m³ for hydrazine, MMH and UDMH respectively.

Hydrazine and both of the derivatives are miscible in all proportions with water. Not only are they miscible, they are extremely non-ideal and fit the definition of "hygroscopic", that is, they very readily take up water, and the equilibrium partial pressure of water vapor over the solutions is much lower than that predicted by Raoult's law. Some limited vapor-liquid equilibrium data have been published (5, 6), and neither the hydrazine and water system nor the UDMH-water system obey the Marqules equation (7, 8). This non-ideality is with respect to the liquid solutions. There are no indications of any particular non-ideality of the vapor mixtures.

The three hydrazine species each have somewhat different physical properties, with, in most cases, the properties of MMH being intermediate between those of hydrazine and UDMH. Most of the relevant physical properties of the pure liquids have been collected into a single publication (9).

Computational Methods:

The work of MacKay (10) was used to predict a mass transfer coefficient based on the vapor phase resistance. The basic equation is:

$$k_m = 0.0292 \text{ Sc}^{-0.67} \text{ u}^{(2-n)/(2+n)} \text{ x}^{(-n)/(2+n)}$$
 (1)

where:

km - mass transfer coefficient,

Sc - Schmidt No.

u - Wind velocity (at elevation of 10 m)

x - "Diameter" of Liquid Pool

(The pool "diameter" is actually the distance across the pool in the direction of the wind.)

n - wind velocity profile power

The Schmidt number is a common dimensionless number, encountered frequently in mass transfer and is given by:

where:

- viscosity

p - density

D - diffusivity of diffusing component in air

The appropriate properties for the Schmidt number evaluation is the gas film adjacent to the liquid surface.

The evaporative flux, then is given by:

$$Q_m = k_m(C_1 - C_A) \tag{2}$$

where:

Q = evaporative flux

C1 = equilibrium concentration of diffusing material immediately adjacent to the liquid surface

CA - concentration of diffusing component in the bulk air stream

For any concentration, C = p/RT if the ideal gas low holds, where:

p = partial pressure of the component

R = universal gas constant

T - temperature

It was assumed that each transfering component will transfer in accordance with equation (2), since it is, essentially, a definition of the mass transfer coefficient. For an evaparating hydrazine, C_A , that is, the bulk concentration in the air, is zero at all times, and that the interface composition may be determined by assuming it to be an equilibrium concentration. For water, which may transfer in either direction, the bulk concentration varies with the humidity, while the interface composition will be an equilibrium value. For carbon dioxide, it was assumed that the composition was 0.03 percent in the air and was zero at the interface.

The Schmidt numbers as well as the equilibrium concentrations are functions of temperature, with the latter being very strong functions for each component (except carbon dioxide). The estimation methods are presented in Appendix A.

The temperature immediately adjacent to the liquid surface was assumed to be equal to the liquid pool temperature, while the mean temperature of the film (for Schmidt number evaluation) was assumed to be the mean of the liquid pool temperature and the bulk air temperature

The liquid pool temperature depends upon the relative rates of heat transfer to and from the pool. The transfer to the pool being radiant heating from sunshine, q_s , radiation from the atmosphere, q_a , convective heating from the air, q_h and convective heating from the ground surface, q_g . It is possible that q_h and/or q_g could be negative. The heat losses are reradiation from the pool to space q_r , and the latent heat of vaporizing materials, q_1 .

The latent heat term is quite large, and is itself a strong temperature function since the evaporation rate is a strong temperature function. Therefore, it is reasonable to assume that the actual pool temperature is that which is in equilibrium with the surroundings, that is, the rate of heat gain is equal to the rate of heat loss.

Thus:

$$q_s + q_a + q_h + q_g = q_r + q_1$$
 (3)

Except for q_s and q_a , each term in equation (3) is a function of the pool temperature, and so it is possible to deduce the pool temperature which would be the equilibrium temperature by finding that temperature which will satisfy equation (3).

The evaluation of q_s was done by an algorithm devised to determine the solar insolation rate on horizontal surfaces as a function of latitude, longitude, date, time, and cloud cover (11). The algorithm is rather lengthy and is not reproduced here. The absorptivity of the pool, a_p, was assumed to be 0.96 which compares well with water and a number of organic liquids (10, 12).

The atmospheric long wave length radiation, q_a , was evaluated as a function of the temperature of the air and the atmospheric emissivity, E_a , as given by Sax (13).

The convective transfer from (or to) the air was modelled after MacKay (10) which is a heat transfer analog of the same mass transfer coefficient given earlier as equation (1). The form for the analogous equation is:

where:

p - density of the "film"

Cp - heat capacity of the "film"

Pr = Prandtle no., Cpu/k of the "film"

k = thermal conductivity

The film properties were evaluated at the mean temperature and the mean composition of the interface (equilibrium composition at the pool temperature) and the bulk air condition. The convective transfer between the ground and the pool was treated according to Schmidt and Silveston (14). This is the same algorithm used by Ille and Springer (3), although the liquid mixture properties had to be evaluated as composition functions as well as temperature functions. These properties estimations and others are presented in Appendix A.

The re-radiation from the pool is that of a radiator radiating to space. The liquid pool emissivity, $E_{\rm p}$, is assumed to be 0.96 (12).

Finally, the latent heat of vaporization is the specific latent heat of each component times its evaporative flux. When the several terms of Equation (3) are rewritten as temperature functions, it becomes:

$$q_g T_a^4 \sigma E_a + h_a (T_a - T_p) + h_g (T_g - T_p) = T_p^4 \sigma + \sum_{i=1}^m Q_{m_i}$$
 (5)

where:

o - Steffan-Boltzmann Constant

Each term of this equation is an energy flux.

Equation (5) may be solved for the pool temperature, T_p, if all the other terms are known. The solution requires an iterative method, but is readily found by the Newton-Raphson method.

The mass transfer flux, as given by Equation (2), is seen to be a function of the equilibrium concentration of transfering component. In Equation (2) this concentration is identified as C_1 . For any component, i, this equilibrium composition is given by:

$$(c_1)_i = \chi_i P_{vi} \times_i / RT_p$$
 (6)

where:

Y - activity coefficient

Pv - vapor pressure

x - mole fraction of component in the liquid

The activity coefficient is predicted by methods indicated in Appendix A.

Equation (6) was not applied to carbon dioxide, but rather, the equilibrium composition was set at zero.

The bulk phase composition, CA, was set at zero for the hydrazines, a nominal value (fixed) was chosen for carbon dioxide, and the value for water was computed from the atmospheric conditions as given in Appendix A.

Overall, the computational procedure requires a pool temperature estimate.

In all this work, the first estimate was taken to be the air temperature, Ta. Based upon this estimate, a set of appropriate physical properties was deduced, and an evaporative flux determined. Finally, with estimated properties and estimated flux, Equation (5) could be solved for the pool temperature.

An overall interation followed, based upon revised estimated pool temperatures until satisfactory agreement existed, at which time the corresponding evaporative flux was taken as the correct value, and the total evaporation rate was determined as the product of the flux times the pool area.

Dispersion calculations used the evaporation rate as the source strength for the hydrazine materials. The basic assumptions were that the source was a groundlevel point source with no plume rise, and in case of two toxic materials present simultaneously (aerozine 50), the toxicity was considered additive, but not synergistic after the method of the American Conference of Industrial Hygenists (14).

Any reaction of the evaporated hydrazine as derivatives in air was treated as a first-order decay. The equations for dispersion are as given by Turner (15) and as used in Ille and Springer (3). The basic relation for centerline concentration for any distance x downwind is:

$$C_{(\mathbf{x},\mathbf{o})} = \frac{Q_{\mathbf{m}}A_{\mathbf{p}}}{\pi\sigma_{\mathbf{v}}\sigma_{\mathbf{z}}\mathbf{u}} \exp - (k_{\mathbf{r}}\mathbf{x}/\mathbf{u})$$
 (7)

where:

k. - atmospheric decay constant

Ap - pool area

ov - crosswind diffusion coefficient

o, - vertical diffusion coefficient

At any distance y away from the centerline, the concentration is given by:

$$C_{(x,y)} = C_{(x,o)} \exp - 1/2(\frac{y}{\sigma_y})^2$$
 (7,a)

The diffusion coefficients, σ_y and σ_z are functions of the downwind distance, x. The values used in this work are those given in Ille and Springer (3) and are as recommended by the Environmental Protection Agency.

For the case of more than one toxic mater al, the existence of a hazardous condition may be determined as follows for n toxic components:

$$\frac{C_T}{L_T} = \sum_{i=1}^{n} \frac{C_i}{L_i} \tag{8}$$

where the ratio $\frac{C_T}{L_T}$ - Combined hazard $\sqrt{2}$ 0

Li - Exposure limit for component i

C - Concentration of component i

If the value of $\frac{C_T}{L_T}$ is less than unity, the exposure limit is not exceeded.

In use, the dispersion calculation was carried out by evaluating Equations

(7) and (7,a) at particular sets of x and y values for each toxic component, then evaluating Equation (8) to determine if a hazard existed. Practically, the downwind distance was successively incremented, and at each, the crosswind distance was incremented until the combined hazard ratio was less than unity. The computation was completed when the centerline (y = 0) concentrations (with successive downwind distances) showed a combined hazard ratio of less than unity.

Computational Results

The desired output of the computing program was an identification of the hazard corridor for a particular spill. It was considered that the hazard corridor was defined if the maximum downwind distance and the maximum crosswind distance of the hazard area were determined.

The computed output could have been used to determine the shape of the hazard area, but there are such a large number of variables in the conditions that it seemed appropriate to simply define a length (maximum downwind distance of hazard) and width (maximum crosswind distance of hazard).

The several variables, atmospheric conditions as well as spill dimensions were arbitrarily set for a "base case" for each of the propellant mixtures considered. Then, in order to determine the effects of the several variables independently, a series of computations were made wherein the selected variable was varied incrementally from the base case, and the results determined in the form of "correction factors". The correction factors derived from these calculations are plotted in the figures following. Tabulated values are presented in Appendix E.

Table I defines the "base case" conditions, and Table II shows the results of the base case calculation.

Table I

DEFINITION OF BASE CALE SPILL CONDITIONS

Spill size: 36 m3 (carload)

Spill area: 350 m²

Spill diameter*: 20 m

Air temperature: 25°C

Ground temperature: 25°C

Wind speed: 3 m/s (6.7 miles/hr)

Stability class: (

Relative humidity: 60%

Latitude: 30° North**, Longitude: 100° West**

Date: 266** (September 23), Time: 10:00 a.m.**

Cloud cover: 302**

*Spill "diameter" is actually the distance across the pool in the direction of the wind.

**The date, time, location and cloud cover conditions result in an insolation (solar flux) of 440 w/m².

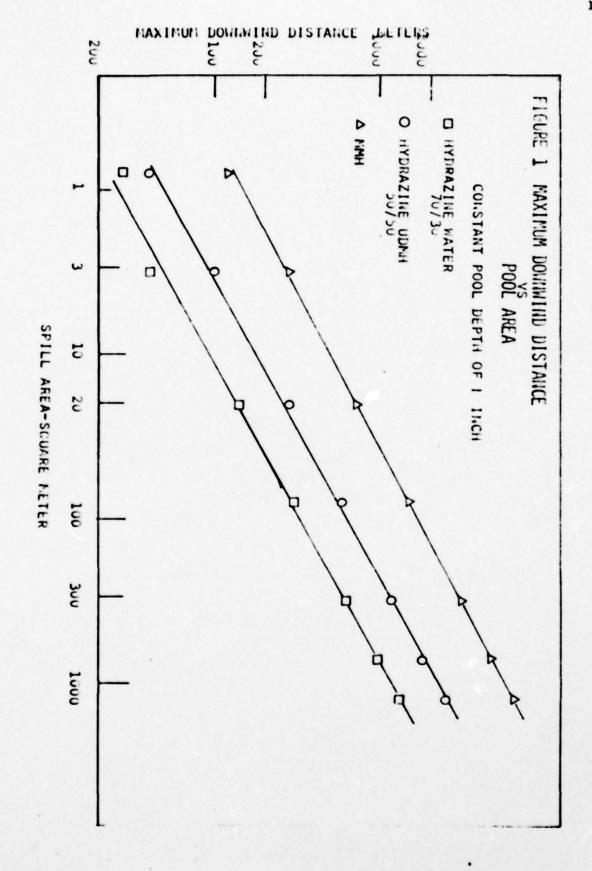
Table II

BASE CASE DISPERSION RESULTS

	Mass	Maximum Downwind	Maximum Crosswind	Amount of Water to
Mixture	of Spill (kg)	Distance of Hazard (m)	Distance of Hazard (m)	Eliminate Hazard* (kg)
Hydrazine: water (70:30)	36070	360	35	216000
Hydrazine: UDMH (50:50)	32620	630	58	780000
MMH "pure"	31330	1630	138	4480000

*The amount of water which would be required to sufficiently dilute the spill so that it would no longer present a hazard was computed by assuming a hazard would no longer exist if the downwind distance of the hazard were no more than the pool diameter.

Since neither the downwind nor crosswind distances are linear in pool area, plots of those distances as functions of pool area are shown in Figures 1 and 2. The figures 3 through 10 are correction factors to the data presented in Figures 1 and 2.



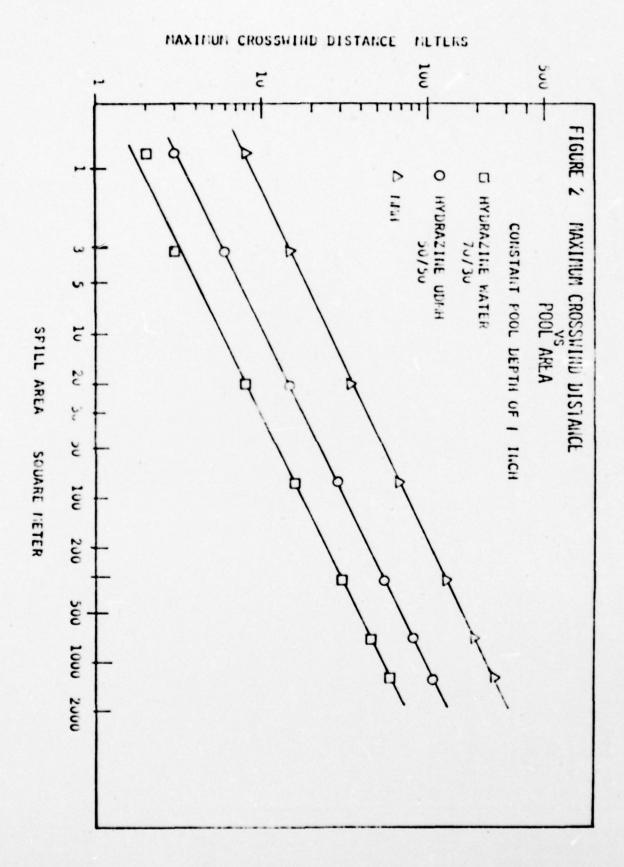


FIGURE 3 CORRECTION FACTORS FOR AIR TEMPERATURE (DOWNWIND AND CROSSWIND)

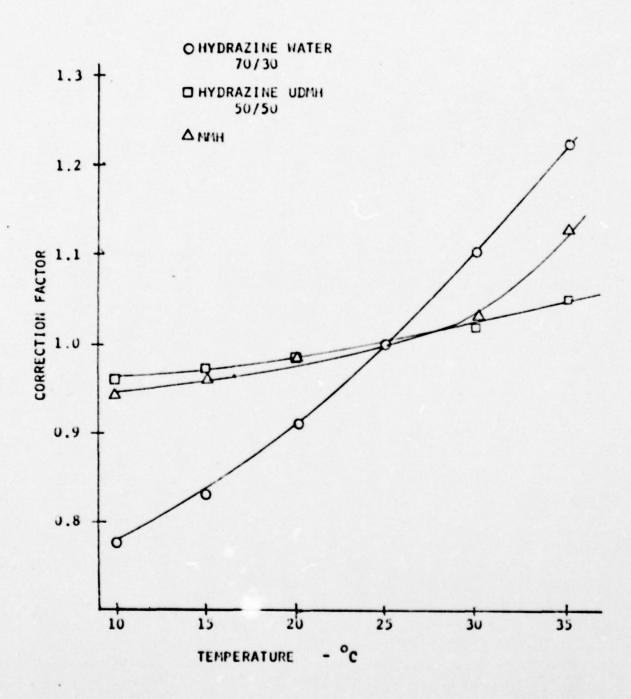


FIGURE 4 CORRECTION FACTORS FOR GROUND TEMPERATURE (DOWNWIND AND CROSSWIND)

O HYDRAZINE WATER
70/30

HYDRAZINE UDNH
50/50

MMH

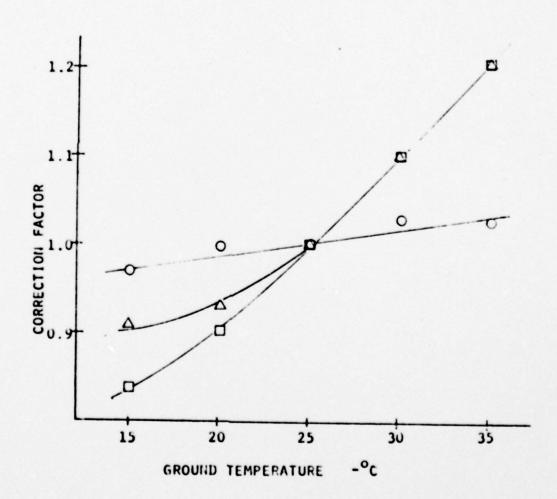


FIGURE 5 CORRECTION FACTORS FOR SOLAR FLUX

(BASE CASE FLUX = 440 W/N2)

APPLIES TO BOTH DOWNWIND AND TO CROSSWIND MAXIMUM DISTANCES

O HYDRAZINE WATER

HYDRAZINE UDNII

HMH A

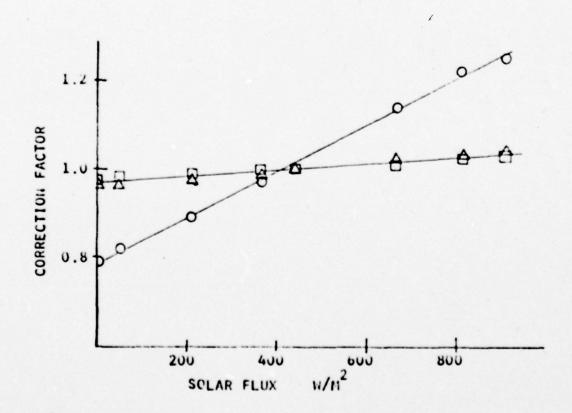
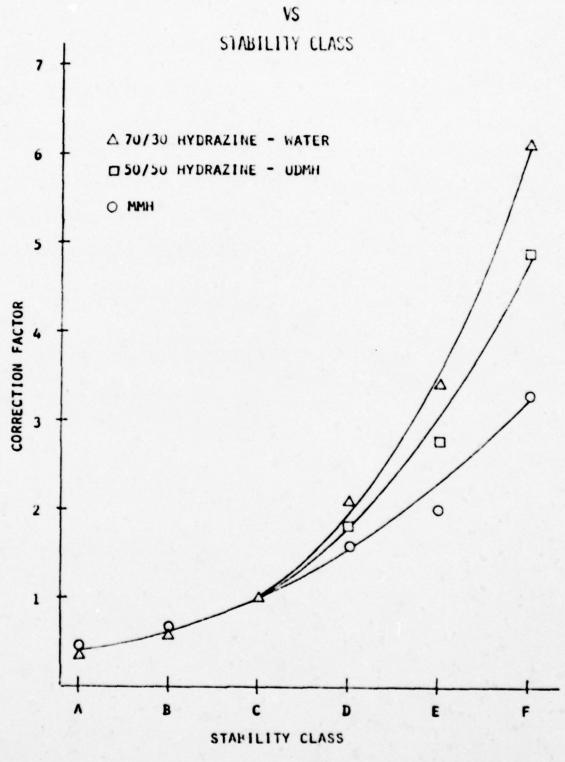


FIGURE 6
CORRECTION FACTOR FOR MAXIMUM DOMINITUD DISTANCE



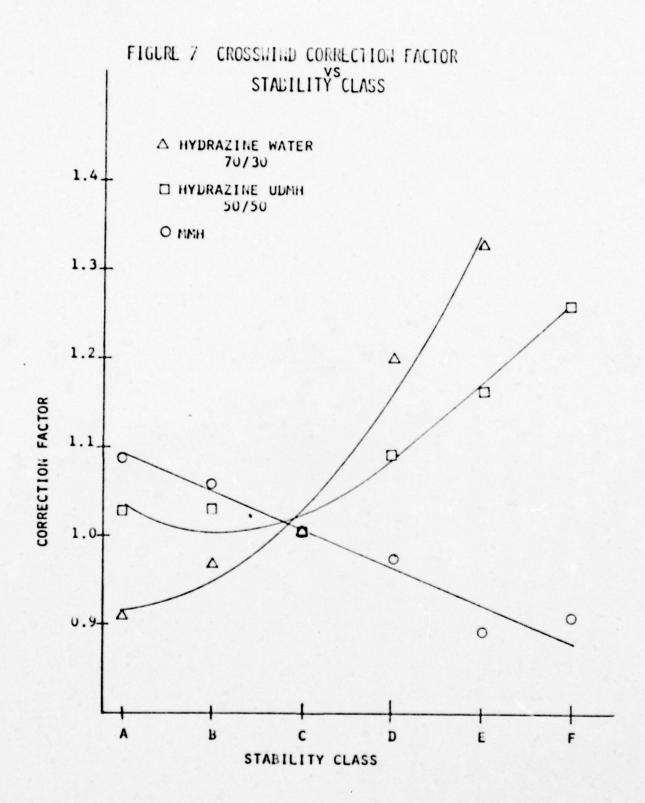


FIGURE 3 CORRECTION FACTORS FOR WIND VELOCITY

CURVE APPLIES FOR ALL MIXTURES, BOTH DOWNWIND AND CROSSWIND

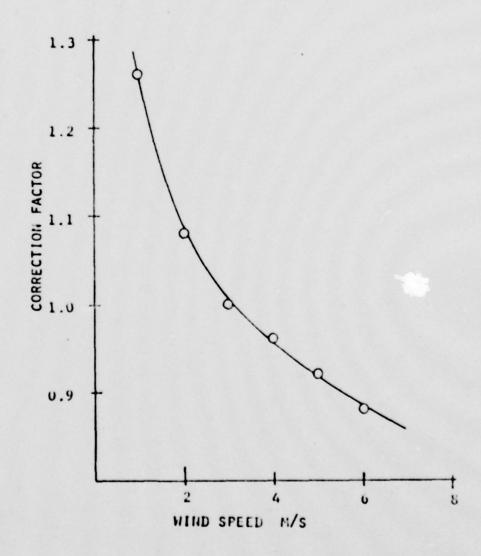


FIGURE 9 CORRECTION FACTORS FOR CLOUD COVER

(DOWNWIND AND CROSSWIND)

O HYDRAZINE WATER 70/30

HYDRAZINE UDMH 50/50

HIMI A

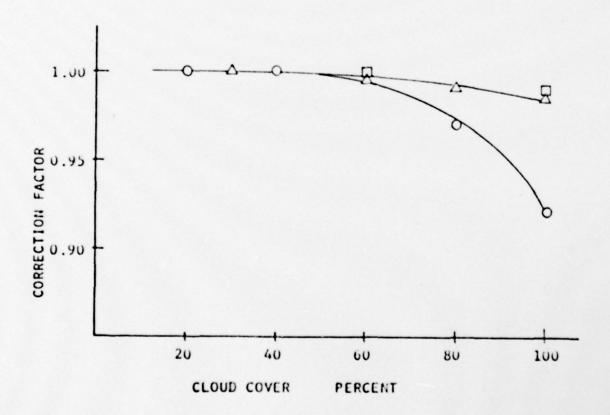
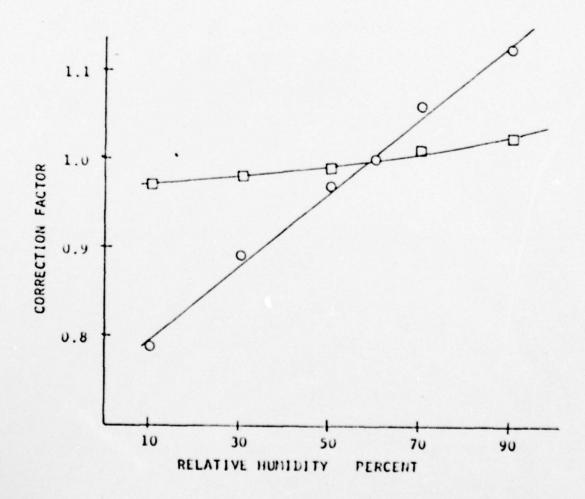


FIGURE 10 CORRECTION FACTORS FOR RELATIVE HUMIDITY (DOMINING AND CROSSWIED)

O HYDRAZINE WATER 70/30

HYDRAZINE UDMH AND MINE



Use of Pre-computed Results

In order to provide a method of rapid field estimation of the extent of a hazard corridor, the maximum distances of hazard presented in Figures 1 and 2 may be multiplied by one or more of the correction factors presented in the following Figures 3 through 10.

Comparisons between the computed result and the estimated result (using the correction factors) have indicated that the maximum difference between computed and estimated results would probably not exceed 20 percent, although all possibilities have not been explored. Also, for small spills, the use of distance increments of ten meters may introduce some additional rounding and truncating errors. An error of 25 percent, if that were the case, would not be an excessive error for such a computation, and could actually be safely ignored when it is compared to the possible error in the dispersion calculations.

Turner (15) points out that the dispersion calculation result will be "within a factor of three" of the actual concentration when the terrain and circumstances agree reasonably well with the assumptions of the model.

An example of the use of the precomputed results follows:

Example case:

Spill of 250 kg of MMH with a spill area of 10 m²

Rel. Humidity = 40%

Cloud Coverage = 0%

Wind Speed = 2 m/s

Stability Class = B

Air Temperature = 30°C

All other conditions similar to "Base Case".

From Fig. 1, the downwind maximum is 250 m

From Fig 2, the crosswind maximum is 21 m

Air temperature correction, Fig. 3, is 1.03

Stability class correction: Fig. 6, Downwind, 0.6; Fig. 7, Crosswind, 0.97

Wind velocity correction, Fig. 8, is 1.08

Cloud cover correction, Fig. 9, is 1.00

Relative humidity correction, Fig. 10, is 0.98

Estimated values:

Downwind max. = 250 x 1.03 x 0.6 x 1.08 x 1.00 x 0.98 = 163 m

Crosswind max. = 27 x 1.03 x 0.97 x 1.08 x 1.00 x 0.98 = 29 m

The computed values for this case are:

Downwind max. - 170 m

Crosswind max = 28 m

Limitations of the Model:

While the model elements which have been assembled represent the state-ofthe art in predicting evaporation, there are several areas of weakness among
the components of the model. Each of these weaknesses are considered below,
although in general it is not possible to evaluate the magnitude of error which
would follow from possible inapplicability of assumptions or models.

Dispersion Estimates

As was mentioned in the preceeding, the dispersion estimation may have errors of substantial magnitude. Turner (15) suggests for level terrain, and for center-line concentrations close to the source, that the estimated value is within a factor of three, which seems to indicate a possible error of some 200 percent.

Compared to this, the other errors in the model are not very significant.

Mass Transfer Model

The mass transfer model, Equation (1), taken from MacKay (10) is intended as a coefficient for a pure material evaporating, or if not pure, a well stirred mixture in the liquid pool. The reason is that the model assumes all the resistance of mass transfer is in the vapor film. It is probable, however, that a considerable liquid phase resistance will be present in most of the practical applications considered in this study; even, perhaps, the pure MMH, since it is hygroscopic and would develop a water rich layer immediately adjacent to the interface as water is absorbed from the atmosphere.

Although it would be difficult to quantify the effect of the error in this instance, it would be most significant for components which evaporate rapidly.

Equilibrium Concentrations

The binary and ternary system vapor-liquid equilibrium data are necessary for predicting the concentration driving force. All the vapor-liquid equilibrium

data which were available for this study were at one atmosphere total pressure of transported component, that is, at the bubble point (boiling point) of the materials. Actually, the partial pressures, and hence the temperatures, will be much lower than that of the data.

To use this data, it was assumed that the activity coefficients were functions only of composition. For the binary system of hydrazine and water, the relation of activity coefficient to composition could be treated as a polynomial. However, for the ternary system of hydrazine-UDMH-water, the binary data had to be fitted to an integrated form of the Gibbs-Duhem Equation, in this case, the Marqules Equation (7, 8) in order to use the ternary form. The binary data did not fit the Marqules Equation well, and so there must be some error in the ternary predictions.

In the case of the MMH-water system, there were no data available at all, and were estimated as being intermediate between the other two binaries, hydrazine-water and UDMH-water.

The error from these assumptions, especially for the system MMH-water could be as high as seventy-five percent.

Other Errors

Most of the remaining errors are in the properties evaluations, such as viscosity, density, thermal conductivity and so forth of the mixtures. These errors will be relatively insignificant because of the much larger uncertainties of the dispersion model, and perhaps, the equilibrium concentrations.

Generally these properties were computed by recognized correlations, and will give results with less than ten percent error.

APPENDIX A

Property Evaluations and Miscellaneous Computational Methods

I. Properties of Liquid Mixtures:

The liquid properties of interest in these computations were:

Density, p

Viscosity, µ

Thermal Conductivity, k

Heat Capacity, cp

Coefficient of Thermal Expansion, B

The methods of computation, including the effect of temperature on the property were generally as recommended by Reid and Sherwood (16).

Density

For a liquid mixture, the volume was assumed to be additive, hence the density was the mass fraction weighted sum, thus:

$$\rho_{\mathbf{m}} = \sum_{i=1}^{n} \Sigma_{i} \rho_{i} \tag{A-1}$$

where: Xi - mass fraction of component i and

Pi - density of component i.

Individual expressions for the density of each component as a function of temperature were obtained as polynomials, either as presented directly or by reduction of published data.

For hydrazine (9)

$$\rho = 1230.78 - 0.62668(T) - 4.5284 \times 10^{-4}(T^2)$$
 (A-1-a)

For UDMH (9)

$$\rho = 1060.41 - 0.77507(T) - 4.8648 \times 10^{-4}(T^2)$$
 (A-1-b)

For MMH (9)

$$\rho = 1150.34 - 0.93949(T)$$
 (A-1-c)

For water (17)

$$\rho = 1054.4 - 0.19241(T)$$
 (A-1-d)

For the above expressions, the density ρ is in kg/m³ and the temperature is in kelvins.

Viscosity

The liquid mixture viscosities were determined from methods presented by Reid and Sherwood (6).

$$log[log(10\mu_m)] = \rho_m[\frac{\sum_{i} \sum_{j} I_j}{\sum_{i} \sum_{j} M_i}] - 2.900$$
 (A-2)

In this expression, µm = mixture viscosity in centipoise

pm = mixture density in g/cm3

xi - mole fraction of component i

Mi - molecular weight of component i

Ii - viscosity constant for component i

for hydrazine, I = 84.8

for MMH, I = 140.4

for UDMH, I = 196.0

for water, I = 59.8

The temperature effect on viscosity was treated by the "Souder's Method" as given in Reid and Sherwood (16) and this is included in the foregoing equation for the viscosity of liquid mixtures.

Thermal Conductivity

Thermal Conductivity of the liquid mixtures was treated by mole fraction weighting, thus:

$$k_{m} = \sum_{i=1}^{m} k_{i} x_{i}$$
 (A-3)

where: ki - thermal conductivity of component i

xi - mole fraction of component i

The individual component conductivities were determined as temperature function polynomials, either as directly presented or by reduction of published data (9, 17).

For hydrazine:
$$k = 0.088142 + 2.742 \times 10^{-3} (T) - 4.677 \times 10^{-6} (T^2)$$
 (A-3-a)

For UDMH:
$$k = 0.28841 - 4.262 \times 10^{-4} (T)$$
 (A-3-b)

For MMH:
$$k = 0.14236 + 9.2048 \times 10^{-4} (T) - 1.9014 \times 10^{-6} (T^2)$$
 (A-3-c)

For water:
$$k = 0.3427 + 9.178 \times 10^{-4} (T)$$
 (A-3-d)

The thermal conductivities are in $w/m \cdot K$ when the temperature, T, is in Kelvins.

Heat Capacity

The heat capacity values of the liquid mixtures were computed as mass weighted averages, thus:

$$c_{\mathbf{p_m}} = \sum_{i=1}^{m} c_{\mathbf{p_i}} \mathbf{x_i} \tag{A-4}$$

where: Cp; = heat capacity of component i

Xi = mass fraction of component i

The component heat capacities were computed from temperature polynomials, either as directly presented or as deduced by data reduction (9, 17)

For hydrazine:
$$C_p = 1234.8 + 8.4488(T) - 7.7651 \times 10^{-3}(T^2)$$
 (A-4-a)

For UDMH:
$$C_p = 1703.3 + 3.6978(T)$$
 (A-4-b)

For MMH:
$$C_p = 2731.3 - 0.072316(T) + 2.4745 \times 10^{-3}(T^2)$$
 (A-4-c)

For Water:
$$C_p = 2512 + 6.147(T)$$
 (A-4-d)

where: Cp = heat capacity, J/kg·K

T - temperature, K

Coefficient of Thermal Expansion

The coefficient of thermal expansion was computed by noting the change in density with temperature:

$$\beta = \frac{\rho_1^2 - \rho_1^2}{2(T_1 - T_2)\rho_1\rho_2} \tag{A-5}$$

where: ρ_1 , ρ_2 = liquid mixture density at temperatures of T_1 and T_2 respectively.

This function was computed each time needed by finding the density at two different temperatures in the temperature region of interest.

II. Properties of Vapor Mixtures:

The vapor properties necessary for computation of the properties of the air/liquid surface film included;

density

viscosity

heat capacity

thermal conductivity

diffusivity

Schmidt no.

For the hydrazines, the basic information on vapor phase viscosity, heat capacity and thermal conductivity was taken as polynomials from the source previously cited (9). For water, these properties were generally taken from Kern (17). Additionally, the vapor film properties are predominately those of air.

Air properties were taken generally from Perry (12).

The density was computed by assuming the validity of the ideal gas law.

Viscosity

The vapor phase component viscosities, μ_{V} , were computed from the following expressions:

For hydrazine:
$$\mu_V = \frac{1.425 \times 10^{-6} (T)^{1.5}}{(T + 569.478)}$$
 (A-6-a)

For UDMH:
$$\mu_V = \frac{1.1155 \times 10^{-6} (T)^{1.5}}{(T + 493.24)}$$
 (A-6-b)

For MMH:
$$\mu_V = \frac{1.2313 \times 10^{-6} (T)^{1.5}}{(T + 530.23)}$$
 (A-6-c)

For water:
$$\mu_v = 33.333 \times 10^{-8} (T) - 1 \times 10^{-7}$$
 (A-6-d)

For air:
$$\mu_V = \frac{1.45 \times 10^{-6} (T)^{1.5}}{(T+116)}$$
 (A-6-e)

The mixture viscosity was then computed by the following:

$$\mu_{V_{m}} = \frac{\sum_{i=1}^{n} \mu_{V,i} y_{i}(M_{i})^{.5}}{\sum_{i=1}^{n} y_{i}M_{i}^{.5}}$$
(A-7)

where: µv,i = vapor phase viscosity in kg/m·5

yi - mole fraction of component i in vapor

Mi - molecular weight of component i

T - temperature in Kelvins

Heat Capacity

Vapor phase heat capacities were computed by polynomial expressions of the form $C_{p_i} = a_i + b_i T + C_i T^2$. For the components, the following constants were used:

Component	<u>a</u>	<u>b</u>	<u>c</u>
hydrazine	1493.7	3.3133	1.0209×10^{-3}
UDMH	264.43	4.548	-1.5146×10^{-3}
мен	355.22	4.1338	-1.34725×10^{-3}
Water	1656.9	0.61297	1.0657×10^{-3}
air	970.69	0.067864	1.6569×10^{-3}

The heat capacity of the mixture, then was:

$$C_{\mathbf{p_m}} = \sum_{i=1}^{m} y_i C_{\mathbf{p_i}} \tag{A-8}$$

The above constants give heat capacity values in J/kg·K when temperatures are in Kelvins.

Thermal Conductivity

The vapor phase thermal conductivity was estimated by methods recommended by Reid and Sherwood (16), as viscosity and heat capacity functions. The component thermal conductivities were computed by the Eucken approximation as given by Perry (12).

$$k_i = M_i(C_{p_i} + 10393/M_i)$$
 (A-9)

where M₁ is the component molecular weight. For the mixture, the thermal conductivity was computed by:

$$k_{m} = \frac{\sum_{i=1}^{m} y_{i} k_{i} M_{i}^{1/3}}{\sum_{i=1}^{m} y_{i} M_{i}^{1/3}}$$
(A-10)

Diffusivity and Schmidt Number

The values for diffusivity of each component in air, and the corresponding Schmidt number were computed by methods given by Sherwood, Pigford and Wilke (18). These were precalculated as temperature functions for all the components of interest, so in use, the component and the temperature was all that was necessary to compute a value for Schmidt number or the binary diffusivity. No allowance was made for the mixture having more than two components. Each diffusivity, each Schmidt number, and hence, each mass transfer coefficient was determined as if it were the only component being transfered.

III. Computation of Mass Transfer Flux in the Absence of Wind.

In order to account for a small mass transfer effect that would occur in the absence of wind, Equation (1) had to be modified since it can, as written, account only for the turbulent transport.

Sherwood, et.al. (18) present a relation for predicting flux at the surface of a sphere under molecular diffusion. This relation can be rearranged to give a static mass transfer coefficient, ks of:

$$k_{s} = \frac{DP}{RTrP_{RM}}$$
 (A-11)

where: D = diffusivity,

P - pressure,

R - gas constant,

T - temperature, and

r - radius of the sphere.

PRM - mean effective pressure of inert

The pool surface was likened to the surface of a sphere, and the equivalent radius, in terms of the pool area was found. The resulting expression is:

$$k_{s} = \frac{DP}{P_{RM}} \sqrt{\frac{2\pi}{A_{p}}}$$
 (A-12)

where A_p is pool surface area. This expression for static transfer was added to the expression for turbulent transport given in Equation (1).

IV. Computation of Equilibrium Vapor Compositions

The basic equation used to compute vapor equilibrium composition was:

$$y_{i} = \int_{i} X_{i} P_{v,i} / F$$
 (A-13)

where: yi - vapor phase mole fraction of component i

 χ_i = activity coefficient of component i

X₁ - Required phase mole fraction of component i

Pv.1 = vapor pressure of component i

P - total pressure

The activity coefficient is a function of the liquid phase composition.

The equation used in this work was the three-suffix Marqules equation. For a binary mixture:

$$\log Y_1 = x_2^2[A_{12} - 2x_1(A_{21} - A_{12})]$$
 (A-14,a)

$$\log Y_2 = x_1^2[A_{21} - 2x_2(A_{12} - A_{21})]$$
 (A-14,b)

The coefficients for a ternary mixture can be deduced from the binary data if the constants A_{12} , A_{21} , A_{13} , A_{31} , A_{23} and A_{32} are known, in which case:

$$\log Y_1 = x_2^2 [A_{12} - 2x_1(A_{21} - A_{12})] + x_3^2 [A_{13} + 2x_1(A_{31} - A_{13})]$$

$$+ x_2 x_3 [1/2(A_{21} + A_{12} + A_{31} + A_{13} - A_{23} - A_{32})$$

$$+ x_1(A_{21} - A_{12} + A_{31} - A_{13}) + (x_2 - x_3)(A_{23} - A_{32})]$$

For Y_2 , the subscripts are rotated, 1 becoming 2, 2 becoming 3 and 3 becoming 1. For Y_3 , the subscripts are rotated once more.

Nomenclature

a - heat capacity constant

Ap - area of liquid pool

A12, A21, A13, A31, A23, A32 - binary activity coefficient constants

h - heat capacity constant

C - vapor phase concentration in bulk air

C1 - vapor phase concentration at interface

Cp - heat capacity at constant pressure

C(x,y) - ground level concentration resulting from dispersion

D - diffusivity

ha - heat transfer coefficient between air and pool

hg - heat transfer coefficient between ground and pool

I - viscosity estimation constant

k - thermal conductivity

k - mass transfer coefficient

kr - reaction rate constant

m - number of components

n - wind velocity profile power

P = pressure

PRM - mean pressure of non-diffusing components

Pv - vapor pressure

Pr - Prandtl No.

q - heat flux from atmospheric radiation

q. - heat flux from convective exchange between pool and ground

qn - heat flux from convective exchange between pool and air

q1 - latent heat of vaporizing or condensing materials, expressed as flux

qr = reradiation heat flux from pool

qs = solar flux

Qm = evaporative flux

r - radius of sphere equir to pool

R - universal gas constant

Sc - Schmidt number

T - temperature

Ta - temperature of the air

Tg - temperature of the ground

Tp = temperature of the pool

u - wend velocity

X - distance across pool, also mole fraction in liquid

x - mass fraction in liquid

y - mole fraction in vapor, also distance away from centerline

B - coefficient of expansion

8 - activity coefficient

u - viscosity

p = density

o - Steffan-Baltzman

σ_y - crosswind diffusion coefficient

 σ_z - vertical diffusion coefficient

Subscripts (except as otherwise noted above)

1 - component 1

1 - liquid

m - mixture

v - vapor

APPENDIX B

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APPENDIX C

Description of the Computer Program

A computer program was written (see Appendix D) to estimate the evaporation flux and the ground level concentrations which result from the dispersion. Figure 11 shows a simplified flow diagram.

The essential feature of the model is that when the temperature of the pool has been satisfactorily estimated, the flux can be readily deduced. However, since the evaporation flux has a significant bearing on the pool temperature, an iterative solution is indicated.

The program makes an initial assumption that the pool temperature is the same as the air temperature. After this assumption, all the properties and coefficients are evaluated and the evaporative flux computed. When the flux is computed, the energy balance is solved for the pool temperature using the Newton-Raphson method. If the new solution for the pool temperature is used immediately as a revised estimate, then under some conditions the overall solution diverges. Therefore, a damping factor is used to obtain a new temperature assumption.

When the solution has converged satisfactorily, then the last value of the evaporative flux is taken as the correct value and the program proceeds to compute an evaporation rate.

The evaporation rate is assumed constant for one second, and the amount of each component transfering in the one second period is subtracted from the pool inventory, so a new pool composition may be computed.

After each one second of evaporation, a new composition is used. The computation proceeds by summing the one second amounts for 36 seconds (0.01 hour). The average rate over the 36 seconds is then used for the dispersion calculations as the source strength.

For each ten meters down wind, the dispersion subroutise computes the groundlevel concentration at the centerline and at incremental distances away from the centerline until the result shows the concentration to be below the hazard level.

That is, the downwind distance is incremented by ten meters, then the crosswind distance is incremented and so forth, until finally the centerline concentration is found to be below the hazard level.

Various output information is available; the actual output can be whatever is desired.

Among the variables computed which would be of interest as output are:

Crosswind distance of the hazard as a function of downwind distance;

Maximum crosswind distance of the hazard;

Maximum downwind distance of the hazard;

Evaporation rate of each component;

Liquid pool composition.

All of the above are computed for each 0.01 hour

The input data required are:

The composition of the original spill, mass fraction of hydrazine, UDMH, water and MMH;

The mass (kg) of the spill, the "diameter" (downwind distance across the pool) of the pool (m) and the pool area (m²);

The time zone (from Greenwich), latitude, longitude (degrees), date,

(Julian day), time (24 hr, decimal hours), the air temperature (°C),

ground temperature (°C), wind velocity at 10 m elevation (m/s), relative humidity (percent), cloud coverage (percent), and the stability

class integer, (1, 2, 3, 4, 5, 6 for A, B, C, D, E, F, respectively).

Since the program computes the evaporation and dispersion for one hour, new weather data may be read in for succeeding hours up to 24.

Subroutines

The main program calls the following subroutines as required:

- Subroutine SOILH: Returns ground-to-pool heat transfer coefficient as a function of the pool composition, amount, area, temperature and the soil temperature.
- Subroutine <u>SKY</u>: Returns long wavelength radiation flux and the partial pressure of water vapor from the atmosphere as functions of the air temperature, relative humidity, and cloud cover.
- Subroutine SOLAR: Returns solar flux intensity as a function of date, time, latitude, longitude and cloud cover.
- Subroutine AIRH: Returns air to pool heat transfer coefficient as a function of mass transfer coefficients, Schmidt numbers, vapor composition, air temperature and pool temperature.
- Subroutine SCMIT: Returns Schmidt numbers and diffusivities for the transfered components in air as functions of the temperature of the pool and of the air.
- Subroutine GAMMA: Returns equilibrium partial pressures for all transfered components as functions of composition and temperature of the pool.
- Subroutine DISPER: Computes and prints the results of dispersion calculations.

 The input is the evaporation rate, which is the source strength, and weather data.

Appendix D contains a listing of the program and a sample of the output.

Variables Used in Computer Program

Variable Name	Variable Description
X11, X12, X13, X14	initial composition mass % of hydrazine, UDMH, water, and MMH, respectively
MASS	mass of the propellant spill, kg
DIA	diameter of the propellant spill, meters (downwind distance)
AREA	area of the spill, m ²
DATE	Julian day (1 to 366)
TIME	time, hours after midnight (Oto 24)
TA	air temperature, °C (sometimes K, internally)
TG	ground temperature, °C (sometimes K, internally)
U	wind speed, m/s
RHUM	relative humidity, percent
KLAS	atmospheric stability class, numerical, 1 = A, 2 = B, etc.
MPRINT	output printing scheme variable
DELT	time constant in seconds used in calculations
EMIS	pool emissivity
GN	ground roughness factor and temperature factor in the atmosphere
POOLAB	pool absorptivity
PT	atmospheric pressure, Pa
TIN	initial time in hours after midnight
X1, X2, X3, X4	mass fraction of hydrazine, UDMH, water, and MMH, respectively
TP Y	computed temperature of the liquid pool, K
тсс	turbulent contribution to the mass transfer coefficient, m/s

Variable Name	Variable Description
PBM1, PBM2, PBM3, PBM4, PBM5	mean partial pressures of non-diffusing material, Pa
TC1, TC2, TC3, TC4, TC5	mass transfer coefficient for hydrazine, UDMH, water MMH, and CO ₂ , respectively
HVAP1, HVAP2, HVAP3, HVAP4	heat of vaporization for hydrazine, UDMH, water, and MMH, respectively, J/kg.
TPT	temporary pool temperature calculated by using Raphson-Newton method
EVAP1, EVAP2, EVAP3, EVAP4	evaporation flux of hydrazine, UDMN, water, and MMH, respectively
CO2	Diffusing flux of CO2 into the liquid pool, kg mole/m2.s
QVAP	evaporative heat loss of the propellant pool, J/m2 s
CONST	solar flux + radiative heat gain from atmosphere - evaporative heat loss, W/m^2
XM1, XM2, XM4	mole fraction in propellant spill of hydrazine, UDMH, and MMH, respectively
XJ1, XJ2, XJ3, XJ4	weight percent of hydrazine, UDMH, water, and MMH, respectively
SUM1, SUM2, SUM3, SUM4	algebraic sum of the evaporation rates of the respec- tive components over a 36 second time interval, kg/s
AVG1, AVG2, AVG3, AVG4	average evaporation rate over a 36 second time interval for hydrazine, UDMH, water, and MMH, respectively, kg/s
XX	downwind distance, m
YM	crosswind distance, m
CCE	centerline concentration equivalent (dimensionless ratio)
CC1, CC2, CC4	centerline concentration in µg/m³
CCY1, CCY2, CCY4	concentration at YM, µg/m ³
XMDEL	increment of downwind distance, m

Variable Name	Variable Description
RXK1, RXK2, RXK4	reaction rate constants (first order), sec-1
Q1, Q2, Q4	emission rates of components, kg/s
xx	downwind distance, km
SIGZ	dispersion coefficient in vertical direction, m
SICY	dispersion coefficient in horizontal direction, m
ILAST	counting integer, dimensionless
INCR	print frequency increment
TF	average film temperature, K
sc1, sc2, sc3, sc4, sc5	Schmidt number for hydrazine, UDMH, water, MMH, and carbon dioxide, respectively
DIFF1, DIFF2, DIFF3, DIFF4, DIFF5	Diffusivity of hydrazine, UDMH, water, MMH, and CO_2 respectively, in air, m^2/s
GAM(1), GAM(2), GAM(3), GAM(4)	activity coefficient for hydrazine, UDMH, water, and MMH, respectively
VP1, VP2, VP3, VP4	pure component vapor pressure for hydrazine, UDMH, water, and MMH, respectively, Pa
W1, W2, W3, W4	mass of hydrazine, UDMH, water, and MMH, respectively kg
HG	heat transfer coefficient between the ground and the liquid pool, $W/m^2\ K$
Q	total mass of the spill, kg
т	temperature used in temp dependent expressions, K
DN1, DN2, DN3, DN4	liquid density of hydrazine, UDMH, water, and MMH, respectively, kg/m ³
DNLM	density of the liquid mixture, kg/m3
DNMTG, DNMTP	liquid density of the mixture at the ground and pool temperature, respectively, kg/m ³
BETA	expansion coefficient, K-1

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Variable Name	Variable Description
F1, F2, F3, F4, F5	variables related to solar radiation as functions of the date
Z, W	direction cosines of direct solar beam
SALT	solar altitude, radians
CCM	cloud cover modifier
IDN	intensity of direct normal solar radiation, BTU/hr ${\rm ft}^2$
ID	intensity of solar radiation incident on pool surface, Btu/hr·ft ²
SKY	sky diffuse radiation incident on surface, Btu/hr·ft ²
P1, P2, P3, P4	equilibrium partial pressure of hydrazine, UDMH, water, and MMH, respectively, Pa
H	heat transfer coefficient between the liquid pool and the air, $W/m^2 \cdot K$
WM(1), WM(2), WM(3), WM(4), WM(5)	molecular weight of hydrazine, UDMH, water, MMH, and air, respectively
Y(1), Y(2), Y(3), Y(4), Y(5)	average mole fraction in the pool-air film of hydra- zine, UDMH, water, MMH, and air, respectively
DNF	molar density of the vapor film, kg mole/m3
CP(1), CP(2), CP(3), CP(4), CP(5)	gaseous heat capacity of hydrazine, UDMH, water, MMH, and air, respectively in J/kg·K
СРМ	average molar heat capacity of the vapor film, J/kg mole·K
VIS(1), VIS(2), VIS(3), VIS(4), VIS(5)	gaseous viscosity of hydrazine, UDMH, water, MMH, and air, respectively, kg/m·s
VISM	average viscosity of the vapor film, kg/m·s
TK(1), TK(2), TK(3), TK(4), TK(5)	gaseous thermal conductivity for hydrazine, UDMH, water, MMH, and air, respectively, W/m·K
ТКН	thermal conductivity of the vapor film mixture, W/m·K
CPF	average heat capacity of the vapor film mixture, J/kg·K

Variable Name	Variable Description
тс	average mass transfer coefficient of the vapor film mixture, m/s
sc	average Schmidt number of the vapor film mixture
PR	Prandtl number of the vapor film mixture at the film temp

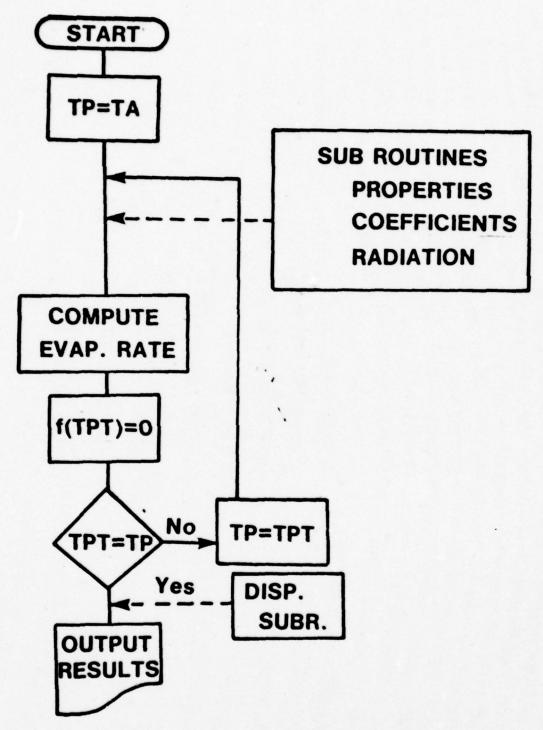


FIGURE 11 PROGRAM FLOW CHART

APPENDIX D

Printout of Computer Program and Typical Output

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APPENDIX E

Computed Results and Correction Factors

MAXIMUM CROSSWIND AND DOWNWIND DISTANCES AS A FUNCTION OF SPILL AREA WITH POOL DEPTH CONSTANT AT 1 INCH (2.54 cm) Table E-1

		(70/3	(70/30) Hydrazine : Water	ne : Water	(50/	(50/50) Hydrazine: UDMH	ne : UDMH	1	100% MMH	
€ (£	Arga (m ²)	Mass (kg)	Crosswind Downwind Distance Distance	Downwind	Mass (kg)	Crosswind Downwind Distance Distance	Downwind Distance	Mass (kg)	Crosswind Distance	Downwind Distance
-	0.79	20	2	14	18	3	20	17.4	80	09
7	3.14	80	3	20	72	9	20	69.5	15	140
s	20	200	80	70	452	15	140	434	35	360
07	79	2000	16	150	1810	53	290	1735	89	750
20	314	8000	31	310	7230	55	290	0769	131	1540
30	710	18000	97	780	16300	82	910	15600	193	2350
9	1260	32000	09	650	28900	107	1230	27800	253	3180
*Base	*Base weather conditions and locale:	lons and lo		1ZN - 6			LATITUD	LATITUDE - 37°N		
			DAT	DATE - 266 Julian day	tan day		LONGITU	LONGITUDE - 100°W		
			MIN	WIND SPEED - 3 m/sec	m/sec		TIME - 10.00	10.00		
			CLO	CLOUD COVERAGE - 30 percent	- 30 per	cent	REL. HU	REL. HUMIDITY - 60 percent	percent	
			AIR	AIR TEMPERATURE = 25°C	E = 25°C		STABILI	STABILITY CLASS: C		
			GRO	GROUND TEMPERATURE = 25°C	TURE = 25	٥.				

Table E-2

SORRECTION FACTORS FOR SOLAR FLUX

(Base Case: SOLR = 440 w/m²)

Solar Flux		ne:Water (/30)	The state of the s	ne: UDMH (/50)		MH (0%)
(w/m ²)	downwind	crosswind	downwind	crosswind	downwind	crosswind
0	0.78	0.80	0.96	0.98	0.96	0.96
49	0.81	0.83	0.98	0.98	0.96	0.96
209	0.89	0.89	0.98	1.00	0.98	0.97
365	0.97	0.97	0.99	1.00	0.99	0.99
440	1.00	1.00	1.00	1.00	1.00	1.00
666	1.14	1.11	1.01	1.02	1.02	1.02
811	1.22	1.20	1.02	1.03	1.05	1.03
909	1.25	1.23	1.04	1.03	1.06	1.05
941	1.28	1.26	1.04	1.05	1.07	1.05
953	1.28	1.26	1.04	1.05	1.07	1.06

Table E-3

CORRECTION FACTORS FOR STABILITY CLASS

Stability		ne:Water /30)		ne : UDMH /50)		MH 90%)
Class	downwind	crosswind	downwind	crosswind	downwind	crosswind
A	0.44	1.09	0.43	1.03	0.33	0.91
В	0.64	1.06	0.63	1.03	0.58	0.97
С	1.00	1.00	1.00	1.00	1.00	1.00
D	1.58	0.97	1.79	1.09	2.11	1.20
E	1.97	0.89	2.75	1.16	3.40	1.33
F	3.28	0.91	4.87	1.26	6.13	1.56

Table E-4
CORRECTION FACTORS FOR WIND VELOCITY

Wind Speed		ne: Water (/30)		ne : UDMH (/50)		MH (0%)
(m/s)	downwind	crosswind	downwind	crosswind	downwind	crosswind
1	1.31	1.29	1.21	1.19	1.28	1.25
2	1.11	1.11	1.06	1.07	1.07	1.07
3	1.00	1.00	1.00	1.00	1.00	1.00
4	0.94	0.97	0.95	0.97	0.94	0.96
5	0.92	0.91	0.90	0.93	0.91	0.92
6	0.86	0.89	0.87	0.90	0.88	0.90

Table E-5
CORRECTION FACTORS FOR AIR TEMPERATURE

Air Temperature	*	ne: Water /30)		ne : UDMH /50)		MH 90%)
(°C)	downwind	crosswind	downwind	crosswind	downwind	crosswind
35	1.25	1.20	1.05	1.04	1.13	1.12
30	1.12	1.09	1.02	1.02	1.03	1.03
25	1.00	1.00	1.00	1.00	1.00	1.00
20	0.91	0.91	0.98	0.99	0.98	0.98
15	0.81	0.85	0.97	0.98	0.96	0.96
10	0.77	0.78	0.96	0.97	0.94	0.95

Table E-6
CORRECTION FACTORS FOR GROUND TEMPERATURE

Ground		ne: Water (30)		ne : UDMH /50)		MH (02)
Temperature (°C)	downwind	crosswind	downwind	crosswind	downwind	crosswind
15	0.97	1.00	0.83	0.84	0.91	0.93
20	1.00	1.00	0.90	0.91	0.92	0.93
25	1.00	1.00	1.00	1.00	1.00	1.00
30	1.03	1.03	1.10	1.10	1.10	1.10
35	1.03	1.03	1.21	1.21	1.21	1.20

Table E-7
CORRECTION FACTORS FOR CLOUD COVERAGE

Cloud Coverage		ne: Water (/30)		ne : UDMH (50)		MH (02)
(1)	downwind	crosswind	downwind	crosswind	downwind	crosswind
20	1.00	1.00	1.00	1.00	1.00	1.00
30	1.00	1.00	1.00	1.00	1.00	1.00
40	1.00	1.00	1.00	1.00	1.00	1.00
60	1.00	1.00	1.00	1.00	0.99	1.00
80	0.97	0.97	0.98	1.00	0.99	0.99
100	0.92	0.91	0.98	1.00	0.98	0.99

Table E-8

CORRECTION FACTORS FOR RELATIVE HUMIDITY

Relative		ne: Water (30)		ne : UDMH (/50)		MH 002)
Humidity (2)	downwind	crosswind	downwind	crosswind	downwind	crosswind
10	0.78	0.80	0.97	0.98	0.95	0.96
30	0.89	0.89	0.98	0.98	0.97	0.98
50	0.97	0.97	0.98	1.00	0.99	0.99
60	1.00	1.00	1.00	1.00	1.00	1.00
70	1.06	1.06	1.00	1.02	1.01	1.01
90	1.14	1.11	1.02	1.02	1.03	1.03